First Principles Study of Mixed Inorganic-Organic Perovskites ($HC(NH_2)_2PbI_3$-$CH_3NH_3PbBr_3$) for Photovoltaic Applications

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To produce low cost and efficient photovoltaic cells, inorganic-organic lead halide perovskite materials appear promising for most suitable solar cells owing to their high power conversion efficiency. Most recent research showes that formamidinium lead iodide ($FAPbI_3$) with methylammonium lead bromide ($MAPbBr_3$) improves the power conversion efficiency of the solar cell to more than 18 per cent under a standard illumination because incorporated $MAPbBr_3$ makes $FAPbI_3$—relatively unstable but comparatively narrow band gap—more stable composition. In respect to first principle study, we investigated band gap of $MAPbI_3$, $FAPbI_3$, $MAPbBr_3$, $(FAPbI_3)_{0.89}$-$(MAPbBr_3)_{0.11}$ and 0.615(eV), 0.466, 1.197, 0.518 respectively through EDISON DFT software. These results emphasize enhancing structure stability is important factor as well as finding narrow band gap.

INTRODUCTION

Dye-sensitized solar cells (DSSCs) are quite an interest in all over the world for its low cost, simple fabrication and high energy conversion efficiency properties. Besides engineering advantages, DSSCs also have distinct characteristics such as transparency, flexibility, colorful allowing practical applications. It is not an overstatement to say that DSSCs lead a solar cell market to next generation. Recently, Perovskite materials integrated into DSSCs was reported by Miyasaka et al. in 2009. Before, production generated only 3.8% power conversion efficiency (PCE) however, according to recent research it reaches 20% PCE allowing the possibility for perovskite solar cells to enter the market.

Specially, inorganic-organic lead halide perovskite materials composed of AMX_3, where A is an organic ammonium cation, M is Pb and X is a halide anion will show promising results.

By using $(FAPbI_3)_{0.85}$-$(MAPbBr_3)_{0.15}$ perovskite solar cell, world record in national renewable energy laboratory (NREL) has been changed.

This research confirmed that incorporation of $MAPbBr_3$ into $FAPbI_3$ stabilizes the perovskite phase of $FAPbI_3$ and improves the power conversion efficiency compared to $FAPbI_3$ or $MAPbBr_3$ each.

Here, we investigate band gap of $MAPbI_3$, $FAPbI_3$, $MAPbBr_3$, $(FAPbI_3)_{0.89}$-$(MAPbBr_3)_{0.11}$ respecitively, and identify that $(FAPbI_3)_{0.89}$-$(MAPbBr_3)_{0.11}$ material's efficiency is related on band gap narrowing or structural stability.

CALCULATION METHODS

We study four perovskites as shown in Fig 1-4 using general gradient approximation (GGA – PBE) in LCAODFTLab of the EDISON Nano-physics that can calculate density functional theory (DFT).

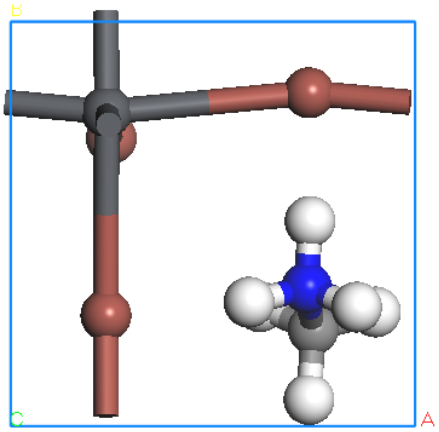

Fig. 40. methylammonium lead iodide

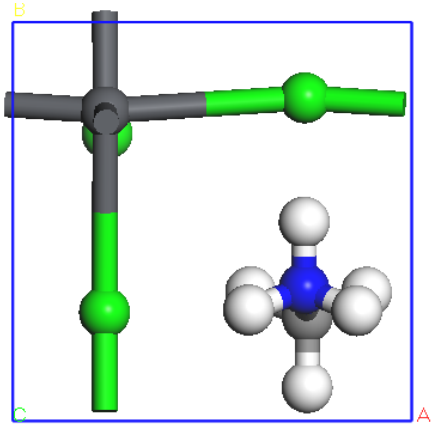

Fig. 41. Methylammonium lead bromide

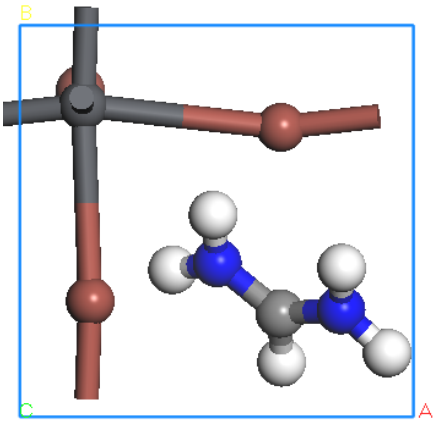

Fig. 42. formamidinium lead iodide

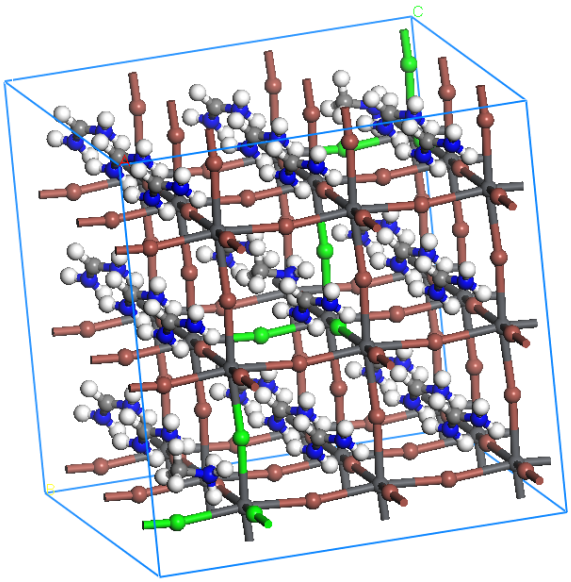

Fig. 43. $(FAPbI_3)_{0.89}$ - $(MAPbBr_3)_{0.11}$

Cubic	$MAPbI_3$	$MAPbBr_3$	$FAPbI_3$	$(FAPbI_3)_{0.89}$ - $(MAPbBr_3)_{0.11}$
Å	6.3425	6.2355	6.4255	19.3485

Table 1. Cubic perovskites lattice constant

We have performed the total energy calculation to find the optimized unit-cell with changing lattice constant.

MAPbI3, MAPbBr3, FAPbI3 perovskite we tested a 5 × 5 × 5 Monkhorst−Pack grid but (FAPbI3)0.89 - (MAPbBr3)0.11 is tested by 1 × 1 × 1 Monkhorst −Pack grid. These were performed with 25 / 200 Ry cutoffs.

RESULTS AND DISCUSSION

We have calculated band structure of 4 perovskites along the M → R → X → Γ → M → Γ. We obtained band gap at R point.

As previously mentioned, $(FAPbI_3)_{0.85}$-$(MAPbBr_3)_{0.15}$ material's efficiency is higher than

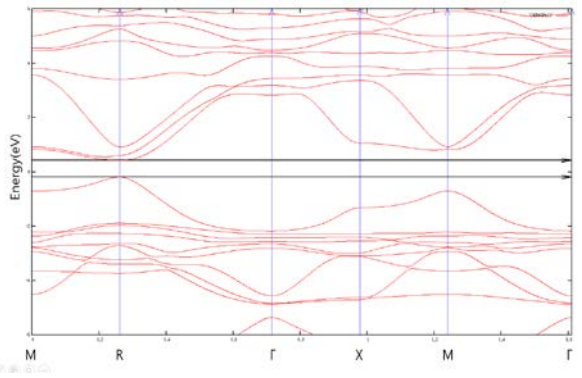

Fig. 44. methylammonium lead iodide band

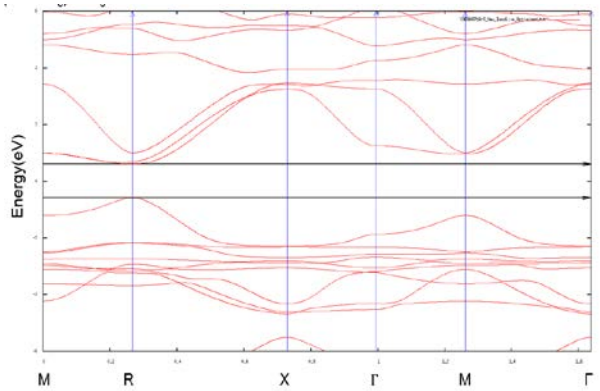

Fig. 45. Methylammonium lead bromide band

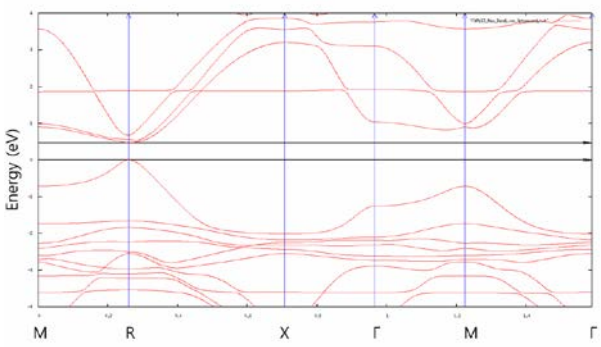

Fig. 46. formamidinium lead iodide band

$MAPbI_3$, $MAPbBr_3$ or FAPbI3. Unfortunately, $(FAPbI_3)_{0.85}$-$(MAPbBr_3)_{0.15}$ model calculation wasn't compatible due to EDISON software's limit. Hence, we substituted the model from $(FAPbI_3)_{0.85}$-$(MAPbBr_3)_{0.15}$ to $(FAPbI_3)_{0.89}$-$(MAPbBr_3)_{0.11}$.

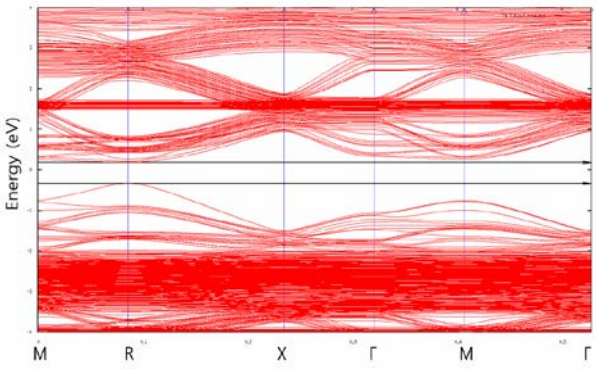

Fig. 8. $(FAPbI_3)_{0.89}$ - $(MAPbBr_3)_{0.11}$ band

Band gap	$MAPbI_3$	$MAPbBr_3$	$FAPbI_3$	$(FAPbI_3)_{0.89}$ - $(MAPbBr_3)_{0.11}$
eV	0.615	1.197	0.466	0.518

Table 2. cubic perovskites band gap

Generally, reduced bandgap may allow absorption of photons over a broader solar spectrum. Focusing only on band gap, $FAPbI_3$ shows high performance between samples. But experimental data doesn't support former assumption.

CONCLUSION

We investigated band gaps of $MAPbI_3$, $MAPbBr_3$, $FAPbI_3$ and $(FAPbI_3)_{0.89}$ - $(MAPbBr_3)_{0.11}$. We study that to achieve high performance perovskite solar cell, consider factors not only narrowing band gap but also such as phase stability, morphology of layer etc.

ACKNOWLEDGEMENT

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REFERENCES

[1] E.Mosconi et al, J. Phys. Chem. C 2013, 117, 13902−13913

[2] A.amat et al, Nano Lett. 2014, 14, 3608−3616

[3] Y.H.Chang et al, Journal of the Korean Physical Society, Vol. 44, No. 4, April 2004, pp. 889~893

[4] S.I.Seok et al, NATURE VOL 517 22 JANUARY 2015